Comment on "Nonclassical Smoothening of Nanoscale Surface Corrugations"

In a recent Letter [1] Erlebacher et al. describe the experimental observation of nonclassical smoothening of a crystalline surface. In their experiment, ripples of typical wavelengths $\lambda_x = 290 - 550nm$ in the x-direction were formed on Si(001) by sputter rippling and then annealed at 650 - 750°C. They report that in contrast with the classical exponential decay with time, the ripple amplitude A(t) followed an inverse linear decay of the type $A(t) = A(0)/[1 + \alpha t]$. Erlebacher et al. interpret their results in terms of a one dimensional step flow model, and compare them with theoretical work of Ozdemir and Zangwill (OZ) [2]. While we have no reservations about the experimental results, we claim that they are inconsistent with the one dimensional step flow model, and a fully two-dimensional model is necessary in order to describe the behavior of the experimental system.

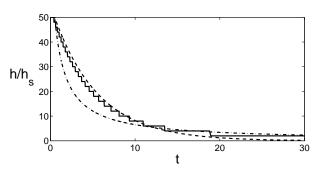


FIG. 1. Amplitude relaxation of an initially sinusoidal grating (solid line) measured in units of the height of an atomic step, h_s . The dashed and dash-dotted lines are fits to an exponential decay and an inverse linear decay, respectively. Time is in arbitrary units.

The mass transport rate limiting process for surface evolution depends on the typical terrace width, Δx , and on the length $\ell = D_s/k$, where D_s is the adatom surface diffusion constant and k is the rate of attachment of adatoms to step edges. The kinetics of the system is diffusion limited (DL) when $\ell \ll \Delta x$ and attachment-detachment limited (ADL) when $\ell \gg \Delta x$. Bartelt et al. showed [3] that for Si(001) surfaces, $\ell \geq 500nm$ for temperatures in the range 670 – 1010°C. This value of ℓ is more than an order of magnitude larger than typical terrace widths in the experiment of Erlebacher et al., and therefore their system should exhibit ADL kinetics.

We studied the relaxation of large-amplitude one dimensional gratings [4] in terms of the same one dimensional step flow model used by OZ [2]. We found that unless the system is purely DL, the height of the grating decays exponentially in time, agreeing entirely with the experiments of Keeffe et al. [5]. Figure 1 shows that even when $\ell = \lambda_x$ and the grating amplitude is 50 steps (values comparable to the experimental ones), an expo-

nential decay fits the step model results better than an inverse linear decay. Thus, the experimental results of Erlebacher et al. are *inconsistent* with the one dimensional theory.

The confusion arises because of the inaccurate statement made by OZ, claiming to have found inverse linear decay in all kinetic limits, but presenting numerical evidence only for DL kinetics. Our work [4] shows unequivocally that the decay is exponential in all cases but purely DL kinetics (which shows inverse linear relaxation).

We now argue that a two dimensional model is needed in order to describe the experiment of Erlebacher et al. The atomic force microscope images in [1] indicate that the corrugations are not purely one dimensional. In addition to the short wavelength ripples along the x-direction, the surface is corrugated in the transverse y-direction with a typical wavelength $\lambda_y \approx 10 \,\lambda_x$. We showed [6] that for radially symmetric morphology the diffusion current J satisfies $J \propto d\mu/d\zeta$, where μ is the local step chemical potential and $\zeta = r + 2\ell h(r)/h_s$, with h_s being the height of a step. The generalization of this relation for non radially symmetric morphology allows an estimate of the order of magnitude of the peak-to-valley directional currents J_x and J_y according to $J_i \propto \Delta \mu/(\lambda_i/2 + 2\ell\Delta h/h_s)$ for i = x or y. Here $\Delta \mu$ is the peak-to-valley chemical potential difference and Δh is the peak-to-valley height difference. According to this, $J_y/J_x \approx 0.95$ in the experiments of Erlebacher et al. A one dimensional model is appropriate only when J_y is negligible compared with J_x , which is clearly not the case here.

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